

CLAIMS

1. A first-order modified, hydrogenated polymer comprising:

5 (1) a hydrogenated polymer obtained by hydrogenating at least one unhydrogenated polymer selected from the group consisting of (1-A) a polymer comprising conjugated diene monomer units and (1-B) a copolymer comprising conjugated diene monomer units and vinyl aromatic hydrocarbon monomer units, said copolymer (1-B)
10 having no or at least one polymer block (H) of said vinyl aromatic hydrocarbon monomer units, and

 (2) a functional group-containing first-order modifier group bonded to said hydrogenated polymer (1),
15 wherein said functional group-containing first-order modifier group comprises at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol group and an alkoxysilane group,

20 said first-order modified, hydrogenated polymer having the following characteristics (i) to (iv):

 (i) a content of said vinyl aromatic hydrocarbon monomer units of from 0 to 60 % by weight, based on the weight of said hydrogenated polymer,

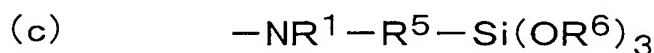
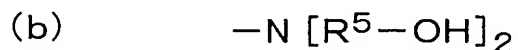
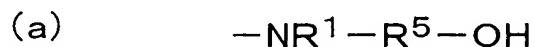
25 (ii) a vinyl aromatic hydrocarbon block ratio of

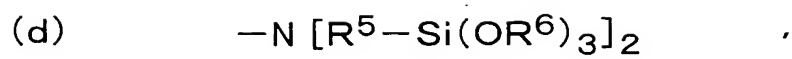
from 0 to less than 50 % by weight, wherein said vinyl aromatic hydrocarbon block ratio is defined as the percent by weight of the vinyl aromatic hydrocarbon monomer units contained in said at least one polymer block (H) of said vinyl aromatic hydrocarbon monomer units, based on the total weight of vinyl aromatic hydrocarbon monomer units contained in said copolymer (1-B),

(iii) a weight average molecular weight of from 20,000 to 2,000,000, and

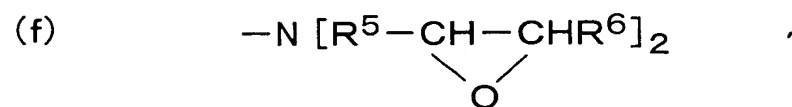
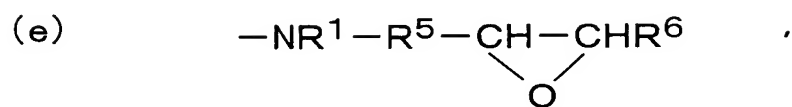
(iv) a hydrogenation ratio of more than 70 %, as measured with respect to the double bonds in said conjugated diene monomer units.

2. The first-order modified, hydrogenated polymer according to claim 1, wherein said functional group-containing first-order modifier group (2) comprises at least one functional group represented by a formula selected from the group consisting of the following formulae (a) to (m):

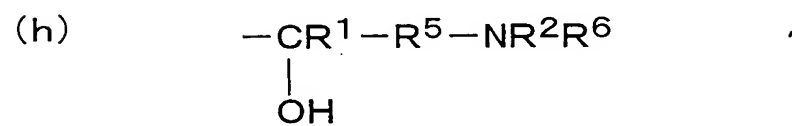
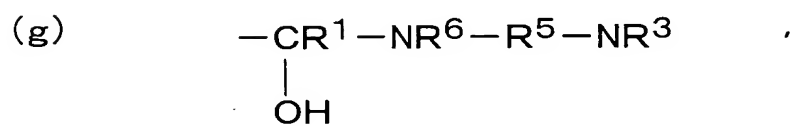




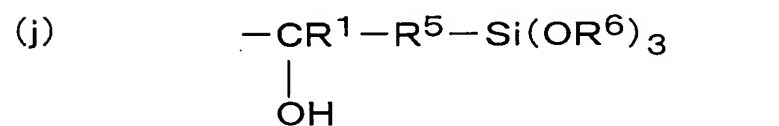
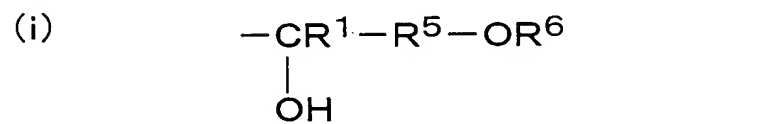
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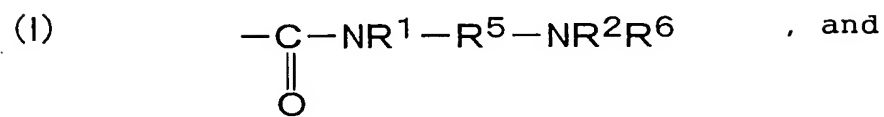
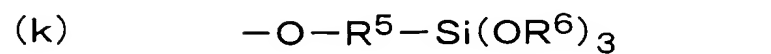
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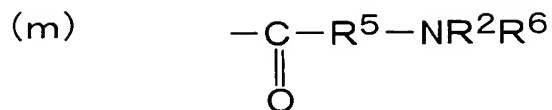
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wherein, in the formulae (a) to (m):

5 N represents a nitrogen atom, Si represents a silicon atom, O represents an oxygen atom, C represents a carbon atom, and H represents a hydrogen atom,

10 each of R^1 to R^4 independently represents a hydrogen atom or a C_1 - C_{24} hydrocarbon group which optionally has at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol group and a C_1 - C_{24} alkoxysilane group,

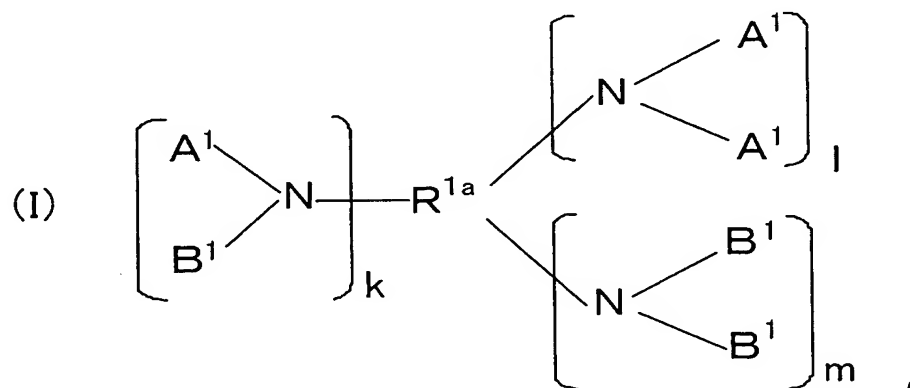
15 each R^5 independently represents a C_1 - C_{48} hydrocarbon group and optionally, independently has at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol group and a C_1 - C_{24} alkoxysilane group,

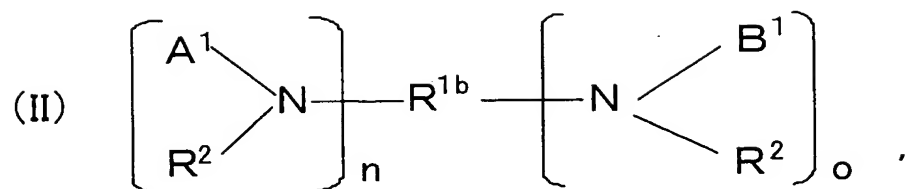
20 each R^6 independently represents a hydrogen atom or a C_1 - C_8 alkyl group,

25 wherein each of R^1 to R^5 optionally, in-

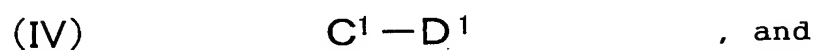
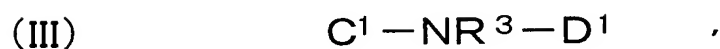
dependently has bonded thereto at least one atom selected from the group consisting of an oxygen atom, a nitrogen atom, a sulfur atom and a silicon atom, said at least one atom being present in a linkage other than a hydroxyl group, an epoxy group, an amino group, a silanol group and an alkoxysilane group.

3. The first-order modified, hydrogenated polymer according to claim 1 or 2, which is represented by a formula selected from the group consisting of the following formulae (I) to (V):

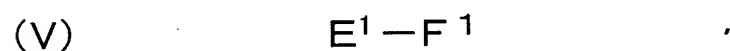




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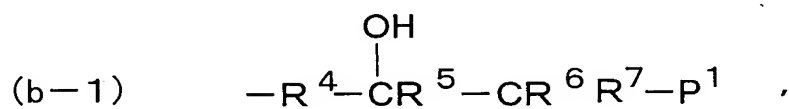
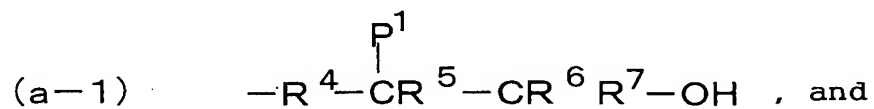
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wherein:

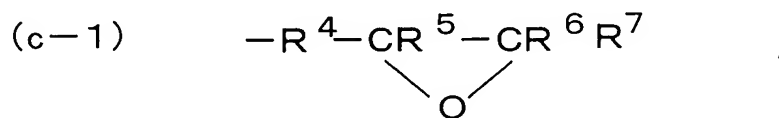
A^1 represents a unit which is represented by any one of the following formulae (a-1) and (b-1):

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B^1 represents a unit which is represented by the following formula (c-1):

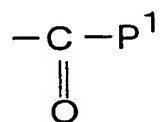


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C^1 represents a unit which is represented by any one of the following formulae (d-1) and (e-1):

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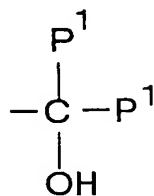
(d-1)



, and

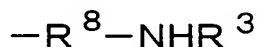
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(e-1)



D^1 represents a unit which is represented by the following formula (f-1):

(f-1)

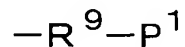


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E^1 represents a unit which is represented by the following formula (g-1):

(g-1)

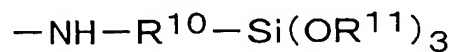


, and

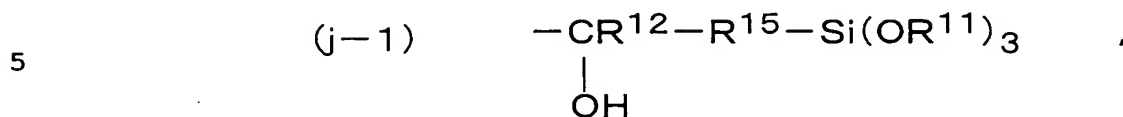
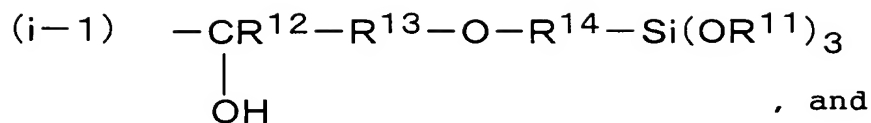
F^1 represents a unit which is represented by any one of the following formulae (h-1) to (j-1):

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(h-1)



,



wherein, in the formulae (I) to (III) and (a-1) to (j-1):

10 N represents a nitrogen atom, Si represents a silicon atom, O represents an oxygen atom, C represents a carbon atom, and H represents a hydrogen atom, P¹ represents said hydrogenated polymer (1),

15 R^{1a} represents a trivalent aliphatic C₁-C₄₈ hydrocarbon group,

each of R^{1b}, R⁴, R⁸ to R¹⁰ and R¹³ to R¹⁵ independently represents a C₁-C₄₈ alkylene group,

20 each of R², R³ and R¹¹ independently represents a C₁-C₄₈ alkyl group, a C₆-C₄₈ aryl group, an alkylaryl group comprised of C₁-C₄₈ alkyl and C₆-C₄₈ aryl, an aralkyl group comprised of C₁-C₄₈ alkyl and C₆-C₄₈ aryl, or a C₃-C₄₈ cycloalkyl group,

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wherein each of R^{1a} , R^{1b} , R^3 , R^4 , R^8 to R^{10} and R^{13} to R^{15} optionally, independently has at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol group and a C_1 - C_{24} alkoxy silane group,

each of R^5 to R^7 and R^{12} independently represents a hydrogen atom, a C_1 - C_{48} alkyl group, a C_6 - C_{48} aryl group, an alkylaryl group comprised of C_1 - C_{48} alkyl and C_6 - C_{48} aryl, an aralkyl group comprised of C_1 - C_{48} alkyl and C_6 - C_{48} aryl, or a C_3 - C_{48} cycloalkyl group,

wherein each of R^{1a} , R^{1b} , R^2 to R^4 and R^8 to R^{15} optionally, independently has bonded thereto at least one atom selected from the group consisting of an oxygen atom, a nitrogen atom, a sulfur atom and a silicon atom, said at least one atom being present in a linkage other than a hydroxyl group, an epoxy group, an amino group, a silanol group and an alkoxy silane group, and

each of k , l , m and o is independently

an integer of 0 or more, provided that both k and l are not simultaneously 0, and n is an integer of 1 or more.

- 5 4. A filler-containing modified polymer composition comprising:

100 parts by weight of (A-1) the first-order modified, hydrogenated polymer of any one of claims 1 to 3, and

- 10 0.5 to 300 parts by weight of (B) a reinforcing filler.

5. The filler-containing modified polymer composition according to claim 4, which further comprises 0.01 to
15 20 parts by weight of (C) a second-order modifier having a functional group which is reactive to said functional group of said modifier group of said first-order modified, hydrogenated polymer (A-1), wherein said second-order modifier (C) is at least one member selected
20 from the group consisting of a functional monomer and a functional oligomer.

6. The filler-containing modified polymer composition according to claim 4 or 5, wherein said reinforcing
25 filler (B) is at least one member selected from the

group consisting of a silica inorganic filler, a metal oxide, a metal hydroxide and carbon.

5 7. A crosslinked, filler-containing modified polymer composition obtained by subjecting the filler -containing modified polymer composition of any one of claims 4 to 6 to a crosslinking reaction in the presence of a vulcanizing agent.

10 8. A modified polymer composition comprising:
1 to 99 parts by weight, relative to 100 parts by weight of the total of components (A-1) and (D), of (A-1) the first-order modified, hydrogenated polymer of any one of claims 1 to 3, and

15 99 to 1 part by weight, relative to 100 parts by weight of the total of components (A-1) and (D), of (D) at least one polymer selected from the group consisting of a thermoplastic resin other than said first-order modified, hydrogenated polymer (A-1) and a rubbery
20 polymer other than said first-order modified, hydrogenated polymer (A-1).

9. The modified polymer composition according to claim 8, which further comprises 0.01 to 20 parts by
25 weight, relative to 100 parts by weight of the total of

components (A-1) and (D), of (C) a second-order modifier having a functional group which is reactive to said functional group of said modifier group of said first-order modified, hydrogenated polymer (A-1),
5 wherein said second-order modifier (C) is at least one member selected from the group consisting of a functional monomer and a functional oligomer.

10. The modified polymer composition according to
10 claim 8 or 9, wherein said rubbery polymer in component (D) comprises at least one member selected from the group consisting of a conjugated diene polymer comprising conjugated diene monomer units, a random copolymer comprising conjugated diene monomer units and vinyl
15 aromatic hydrocarbon monomer units, a block copolymer comprising conjugated diene monomer units and vinyl aromatic hydrocarbon monomer units, a non-diene polymer and a natural rubber,

said rubbery polymer being unhydrogenated or at
20 least partially hydrogenated.

11. The modified polymer composition according to any one of claims 8 to 10, wherein said thermoplastic resin in component (D) is a functional group-containing thermoplastic resin and said rubbery polymer in component
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(D) is a functional group-containing rubbery polymer, wherein each of said functional group-containing thermoplastic resin and rubbery polymer contains at least one functional group which is reactive to said functional group of said first-order modifier group of said first-order modified, hydrogenated polymer (A-1).

12. The modified polymer composition according to claim 11, wherein said functional group-containing thermoplastic resin comprises at least one member selected from the group consisting of a polyester resin, a polyamide resin, a polycarbonate resin, a polyurethane resin, a polyphenylene ether resin and a polyoxymethylene resin each of which contains at least one functional group selected from the group consisting of an acid anhydride group, a carboxyl group, a hydroxyl group, an epoxy group, an amino group, a silanol group and an alkoxysilane group.

13. An adhesive composition comprising:

100 parts by weight of (A-1) the first-order modified, hydrogenated polymer of any one of claims 1 to 3, and

20 to 400 parts by weight of (E) a tackifier.

14. The adhesive composition according to claim 13,
which further comprises 0.01 to 20 parts by weight of
(C) a second-order modifier having a functional group
which is reactive to said functional group of said
5 modifier group of said first-order modified, hydrogen-
ated polymer (A-1), wherein said second-order modifier
(C) is at least one member selected from the group con-
sisting of a functional monomer and a functional oli-
gomer.

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15. An asphalt composition comprising:

0.5 to 50 parts by weight of (A-1) the first-order
modified, hydrogenated polymer of any one of claims 1
to 3, and

15

100 parts by weight of (F) an asphalt.

16. The asphalt composition according to claim 15,
which further comprises 0.01 to 20 parts by weight of
(C) a second-order modifier having a functional group
20 which is reactive to said functional group of said
modifier group of said first-order modified, hydrogen-
ated polymer (A-1), wherein said second-order modifier
(C) is at least one member selected from the group con-
sisting of a functional monomer and a functional oli-
25 gomer.

17. A styrene resin composition obtained by subjecting a raw material mixture to radical polymerization, said raw material mixture comprising:

5 2 to 30 parts by weight, relative to 100 parts by weight of the total of components (A-1) and (G), of (A-1) the first-order modified, hydrogenated polymer of any one of claims 1 to 3, and

 98 to 70 parts by weight, relative to 100 parts by weight of the total of components (A-1) and (G), of (G) a vinyl aromatic hydrocarbon monomer or a mixture of a vinyl aromatic hydrocarbon monomer and a comonomer copolymerizable with said vinyl aromatic hydrocarbon monomer.

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18. The styrene resin composition according to claim 17, wherein said raw material mixture further comprises 0.01 to 20 parts by weight, relative to 100 parts by weight of the total of components (A-1) and (G), of (C) a second-order modifier having a functional group which is reactive to said functional group of said modifier group of said first-order modified, hydrogenated polymer (A-1), wherein said second-order modifier (C) is at least one member selected from the group consisting of

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25 a functional monomer and a functional oligomer.

19. A method for producing the styrene resin composition of claim 17 or 18, comprising:

(1) providing a raw material mixture comprising
5 (A-1) the first-order modified, hydrogenated polymer of
any one of claims 1 to 3, (G) a vinyl aromatic hydro-
carbon monomer or a mixture of a vinyl aromatic hydro-
carbon monomer and a comonomer copolymerizable with
said vinyl aromatic hydrocarbon monomer, and optionally
10 at least one member selected from the group consisting
of (C) a second-order modifier and (B) a reinforcing
filler, and

(2) subjecting said raw material mixture to radi-
cal polymerization,
15 thereby obtaining a styrene resin composition.

20. A second-order modified polymer comprising:

(β) a base polymer, and
(δ) a functional group-containing modifier group
20 bonded to said base polymer (β),

wherein said second-order modified polymer is ob-
tained by reacting a second-order modifier with a
first-order modified polymer comprising (β) a base
polymer and (γ) a functional group-containing first-
25 order modifier group bonded to said base polymer (β) to

thereby form (δ) a functional group-containing modifier group, wherein said second-order modifier has a functional group which is reactive to said functional group of said first-order modifier group (γ) of said first-order modified polymer, and wherein said second-order modifier is used in an amount of 0.3 to 10 moles, relative to one equivalent of the functional group of said first-order modifier group (γ) of said first-order modified polymer,

said second-order modifier being at least one member selected from the group consisting of a functional monomer and a functional oligomer,

wherein said base polymer (β) of said first-order modified polymer is unhydrogenated or at least partially hydrogenated and is at least one member selected from the group consisting of the following polymers (β -1) to (β -3):

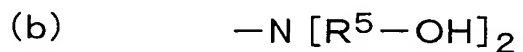
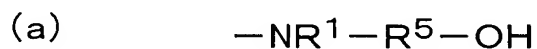
(β -1) a conjugated diene polymer comprising conjugated diene monomer units,

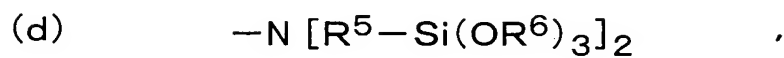
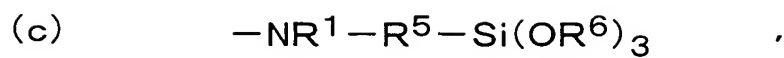
(β -2) a copolymer comprising conjugated diene monomer units and vinyl aromatic hydrocarbon monomer units, said copolymer having no or at least one polymer block (H) of said vinyl aromatic hydrocarbon monomer units, wherein said copolymer has a vinyl

aromatic hydrocarbon block ratio of from 0 to less than 50 % by weight, said vinyl aromatic hydrocarbon block ratio being defined as the percent by weight of the vinyl aromatic hydrocarbon monomer units contained in said at least one polymer block (H) of said vinyl aromatic hydrocarbon monomer units, based on the total weight of vinyl aromatic hydrocarbon monomer units contained in said copolymer as in the unhydrogenated state, and

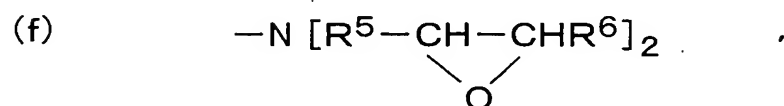
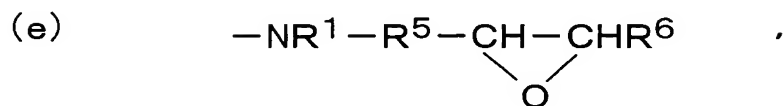
(β -3) a vinyl aromatic hydrocarbon polymer comprising vinyl aromatic hydrocarbon monomer units, and

wherein said functional group-containing first-order modifier group (γ) of said first-order modified polymer comprises at least one functional group represented by a formula selected from the group consisting of the following formulae (a) to (m):

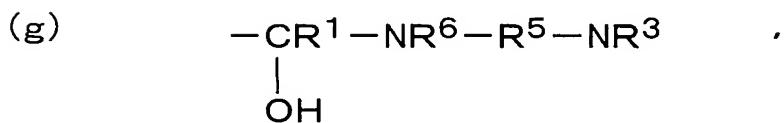




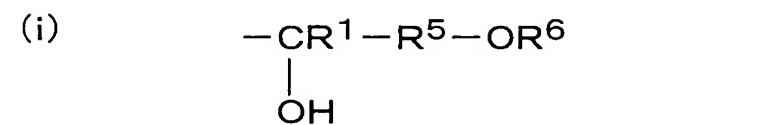
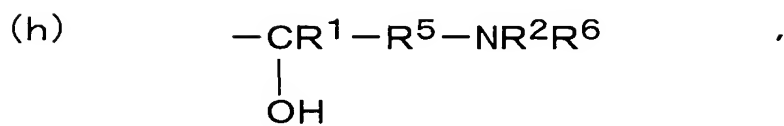
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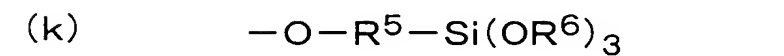
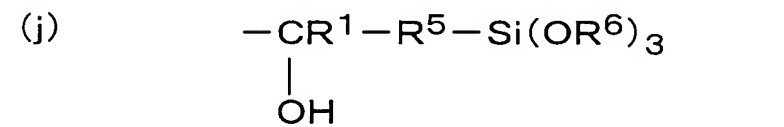
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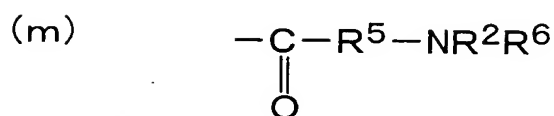
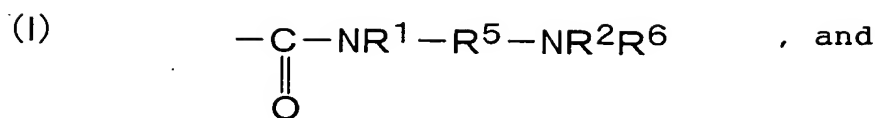
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wherein, in the formulae (a) to (m):

N represents a nitrogen atom, Si represents a silicon atom, O represents an oxygen atom, C represents a carbon atom, and H represents a hydrogen atom,

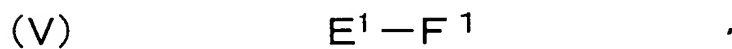
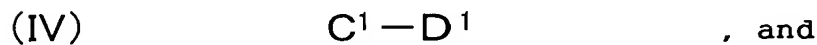
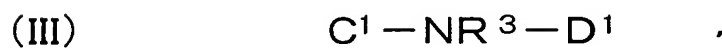
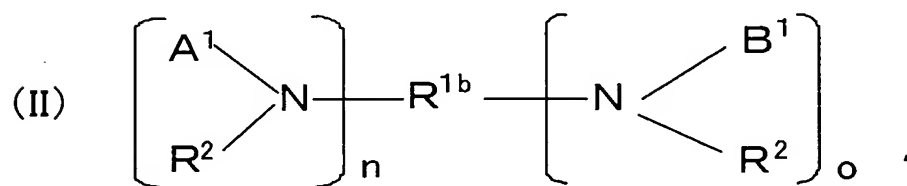
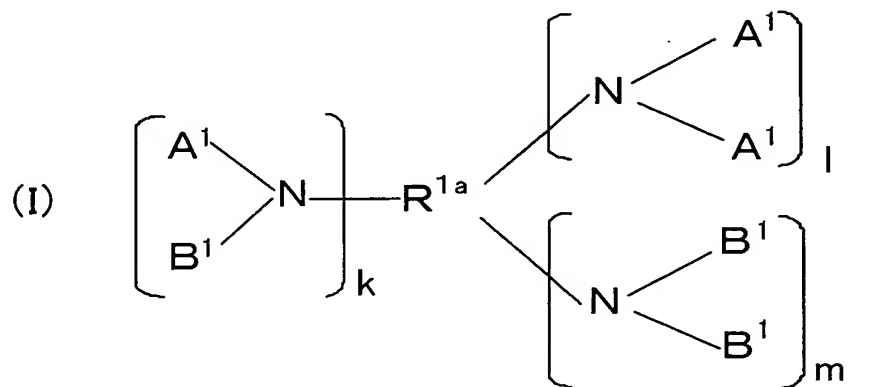
each of R^1 to R^4 independently represents a hydrogen atom or a $\text{C}_1\text{-C}_{24}$ hydrocarbon group which optionally has at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol group and a $\text{C}_1\text{-C}_{24}$ alkoxysilane group,

each R^5 independently represents a $\text{C}_1\text{-C}_{48}$ hydrocarbon group and optionally, independently has at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol group and a $\text{C}_1\text{-C}_{24}$ alkoxysilane group,

each R⁶ independently represents a hydrogen atom or a C₁-C₈ alkyl group,

wherein each of R¹ to R⁵ optionally, independently has bonded thereto at least one atom selected from the group consisting of an oxygen atom, a nitrogen atom, a sulfur atom and a silicon atom, said at least one atom being present in a linkage other than a hydroxyl group, an epoxy group, an amino group, a silanol group and an alkoxysilane group.

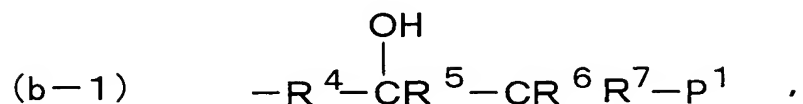
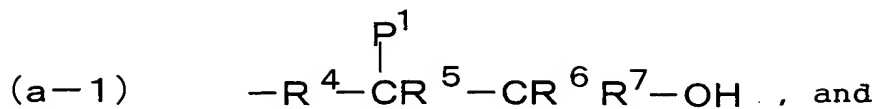
21. The second-order modified polymer according to claim 20, wherein said first-order modified polymer is represented by a formula selected from the group consisting of the following formulae (I) to (V):



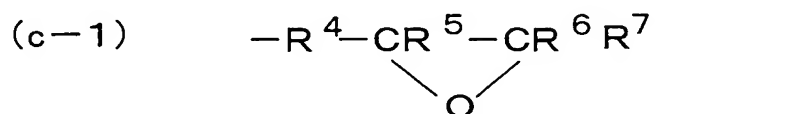
wherein:

A^1 represents a unit which is represented by any

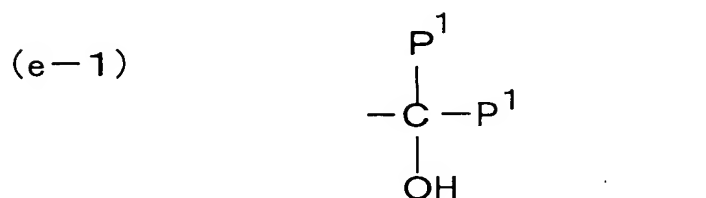
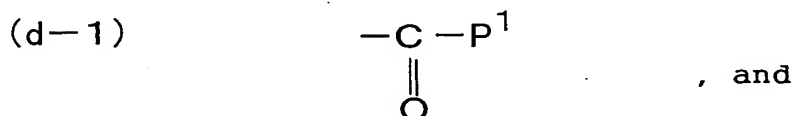
one of the following formulae (a-1) and (b-1):



B¹ represents a unit which is represented by the following formula (c-1):



C¹ represents a unit which is represented by any one of the following formulae (d-1) and (e-1):



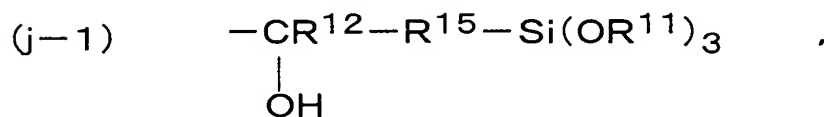
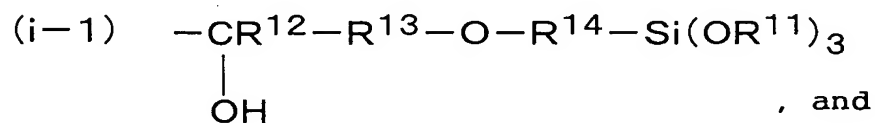
D¹ represents a unit which is represented by the following formula (f-1):



E^1 represents a unit which is represented by the following formula (g-1):



F^1 represents a unit which is represented by any one of the following formulae (h-1) to (j-1):



wherein, in the formulae (I) to (III) and (a-1) to (j-1):

N represents a nitrogen atom, Si represents a silicon atom, O represents an oxygen atom, C represents a carbon atom, and H represents a hydrogen atom,

P^1 represents said base polymer,

R^{1a} represents a trivalent aliphatic C_1 - C_{48} hydrocarbon group,

each of R^{1b} , R^4 , R^8 to R^{10} and R^{13} to R^{15} independently represents a C_1 - C_{48} alkylene group,

each of R^2 , R^3 and R^{11} independently represents a C_1 - C_{48} alkyl group, a C_6 - C_{48} aryl group, an alkylaryl group comprised of C_1 - C_{48} alkyl and C_6 - C_{48} aryl, an aralkyl group comprised of C_1 - C_{48} alkyl and C_6 - C_{48} aryl, or a C_3 - C_{48} cycloalkyl group,

wherein each of R^{1a} , R^{1b} , R^3 , R^4 , R^8 to R^{10} and R^{13} to R^{15} optionally, independently has at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol group and a C_1 - C_{24} alkoxysilane group,

each of R^5 to R^7 and R^{12} independently represents a hydrogen atom, a C_1 - C_{48} alkyl group, a C_6 - C_{48} aryl group, an alkylaryl group comprised of C_1 - C_{48} alkyl and C_6 - C_{48} aryl, an aralkyl group com-

prised of C₁-C₄₈ alkyl and C₆-C₄₈ aryl, or
a C₃-C₄₈ cycloalkyl group,

wherein each of R^{1a}, R^{1b}, R² to R⁴ and
R⁸ to R¹⁵ optionally, independently has
bonded thereto at least one atom se-
lected from the group consisting of an
oxygen atom, a nitrogen atom, a sulfur
atom and a silicon atom, said at least
one atom being present in a linkage
other than a hydroxyl group, an epoxy
group, an amino group, a silanol group
and an alkoxysilane group, and

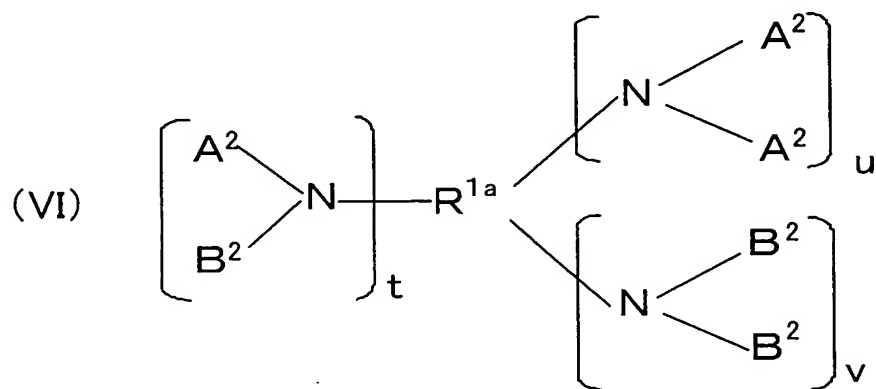
each of k, l, m and o is independ-
ently an integer of 0 or more, provided
that both k and l are not simultaneously
0, and n is an integer of 1 or more.

22. The second-order modified polymer according to
claim 20 or 21, wherein each of said functional monomer
and said functional oligomer has at least one func-
tional group selected from the group consisting of a
hydroxyl group, an amino group, a carboxyl group, an
acid anhydride group, an isocyanate group, an epoxy
group, a silanol group and an alkoxysilane group.

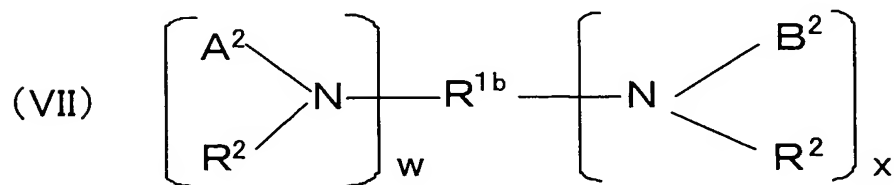
23. The second-order modified polymer according to any one of claims 20 to 22, which is represented by a formula selected from the group consisting of the following formulae (VI) to (X):

5

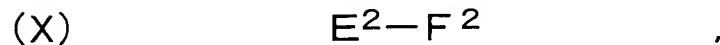
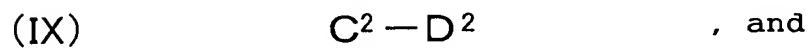
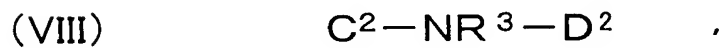
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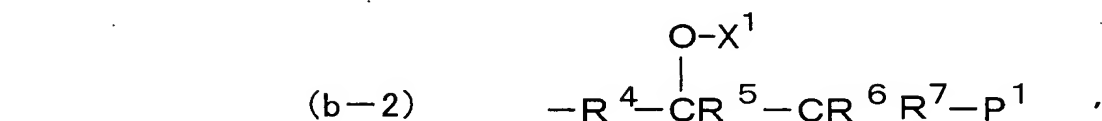
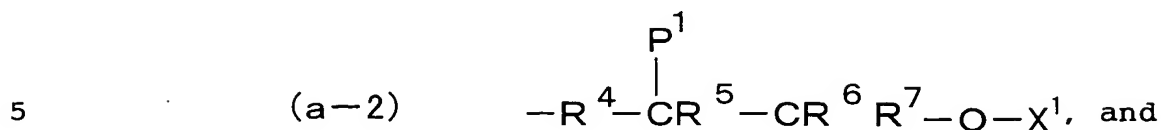
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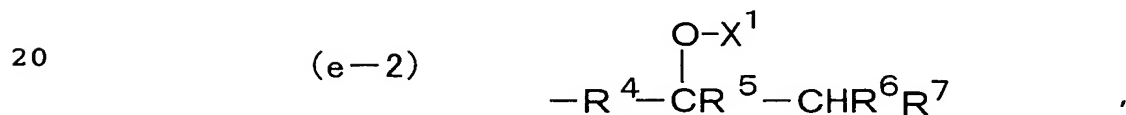
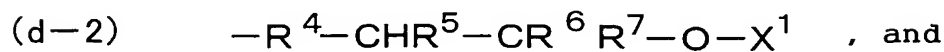
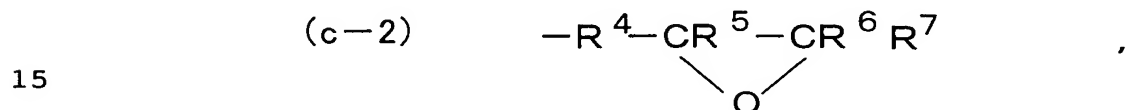
wherein:

A² represents a unit which is represented by any one of the following formulae (a-2) and (b-2):



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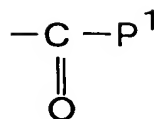
B² represents a unit which is represented by any one of the following formulae (c-2) to (e-2):



C² represents a unit which is represented by any one of the following formulae (f-2) to (h-2):

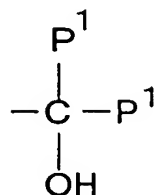
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(f-2)



5

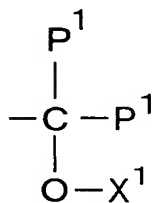
(g-2)



, and

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(h-2)



D² represents a unit which is represented by the following formula (i-2):

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(i-2)



E² represents a unit which is represented by the following formula (j-2):

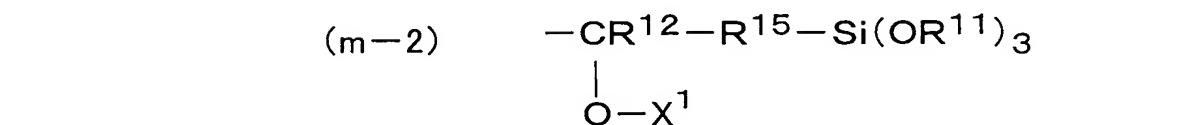
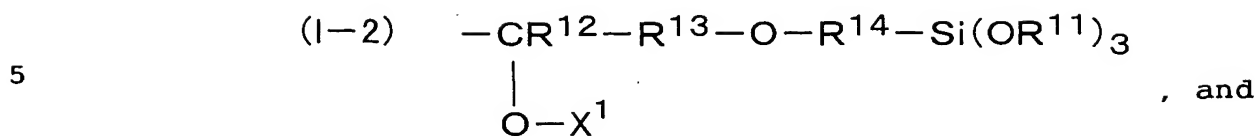
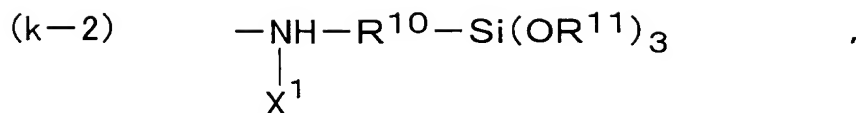
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(j-2)



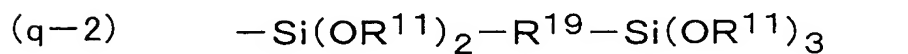
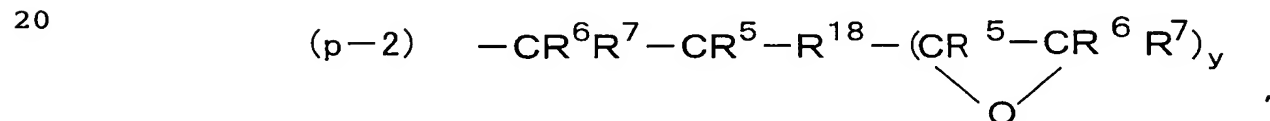
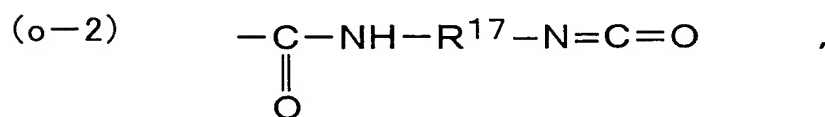
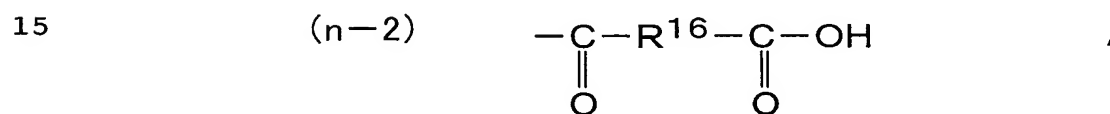
, and

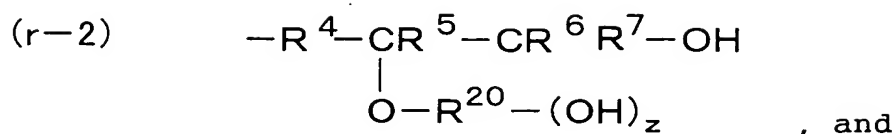
F² represents a unit which is represented by any one of the following formulae (k-2) to (m-2):



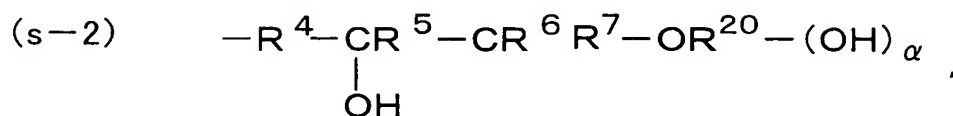
wherein:

X^1 represents a unit which is represented by any one of the following formulae (n-2) to (s-2):





5



10

wherein, in the formulae (VI) to (VIII)
and (a-2) to (s-2):

N represents a nitrogen atom, Si represents a silicon atom, O represents an oxygen atom, C represents a carbon atom, and H represents a hydrogen atom,

15

P^1 represents the base polymer,

R^{1a} represents a trivalent aliphatic C_1 - C_{48} hydrocarbon group,

20

each of R^{1b} , R^4 , R^8 to R^{10} and R^{13} to R^{20} independently represents a C_1 - C_{48} alkylene group,

each of R^2 , R^3 and R^{11} independently represents a C_1 - C_{48} alkyl group, a C_6 - C_{48} aryl group, an alkylaryl group comprised of C_1 - C_{48} alkyl and C_6 - C_{48} aryl, an aralkyl group comprised of C_1 - C_{48} alkyl and C_6 - C_{48}

25

aryl, or a C₃-C₄₈ cycloalkyl group,

wherein each of R^{1a}, R^{1b}, R³, R⁴, R⁸ to R¹⁰, R¹³ to R¹⁵ and R¹⁷ to R²⁰ optionally, independently has at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol group and a C₁-C₂₄ alkoxy silane group,

each of R⁵ to R⁷ and R¹² independently represents a hydrogen atom, a C₁-C₄₈ alkyl group, a C₆-C₄₈ aryl group, an alkylaryl group comprised of C₁-C₄₈ alkyl and C₆-C₄₈ aryl, an aralkyl group comprised of C₁-C₄₈ alkyl and C₆-C₄₈ aryl, or a C₃-C₄₈ cycloalkyl group,

wherein each of R^{1a}, R^{1b}, R² to R⁴ and R⁸ to R²⁰ optionally, independently has bonded thereto at least one atom selected from the group consisting of an oxygen atom, a nitrogen atom, a sulfur atom, and a silicon atom, said at least one atom being present in a linkage other than a hydroxyl group, an epoxy group, an amino group, a silanol group and an alkoxy silane group, and

each of t, u, v and x is independently an integer of 0 or more, provided that both t and u are not simultaneously 0, and each of w, y, z and α is independently an integer of 1 or more.

5

24. A method for producing the second-order modified polymer of any one of claims 20 to 23, comprising:

(1) providing a first-order modified polymer comprising:

10

(β) a base polymer which is unhydrogenated or at least partially hydrogenated and which is at least one member selected from the group consisting of the following polymers (β -1) to (β -3):

15

(β -1) a conjugated diene polymer comprising conjugated diene monomer units,

20

(β -2) a copolymer comprising conjugated diene monomer units and vinyl aromatic hydrocarbon monomer units, said copolymer having no or at least one polymer block (H) of said vinyl aromatic hydrocarbon monomer units, wherein said copolymer has a vinyl aromatic hydrocarbon block ratio of from 0 to less than 50 % by weight, said vinyl aromatic hydrocarbon block ratio being defined as the percent by weight of

25

the vinyl aromatic hydrocarbon monomer units contained in said at least one polymer block (H) of said vinyl aromatic hydrocarbon monomer units, based on the total weight of vinyl aromatic hydrocarbon monomer units contained in said copolymer as in the unhydrogenated state, and

(β -3) a vinyl aromatic hydrocarbon polymer comprising vinyl aromatic hydrocarbon monomer units, and

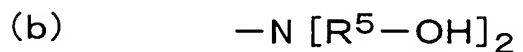
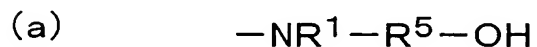
(γ) a functional group-containing first-order modifier group bonded to said base polymer (β),

wherein said first-order modified polymer is produced by a process in which a base polymer having a living terminal is produced by a living anionic polymerization using an organolithium compound as a polymerization catalyst, and a functional group-containing first-order modifier is addition-bonded to said living terminal of said base polymer to obtain a first-order modified polymer, optionally followed by partial or complete hydrogenation of the obtained first-order modified polymer, and

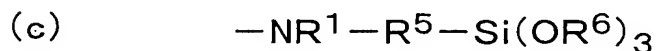
(2) reacting a second-order modifier with said first-order modified polymer to thereby form (δ) a

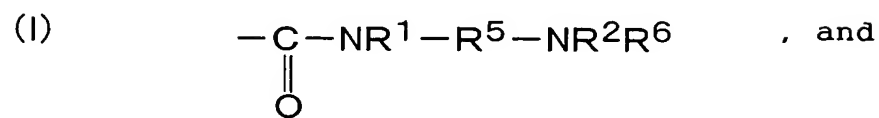
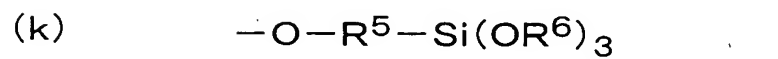
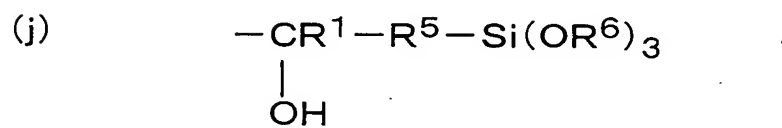
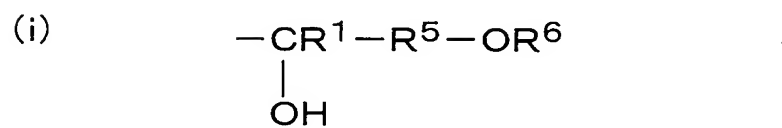
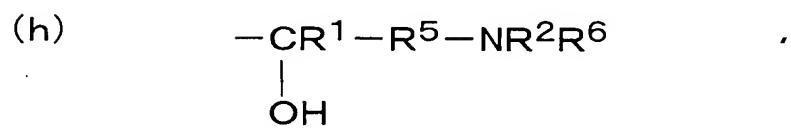
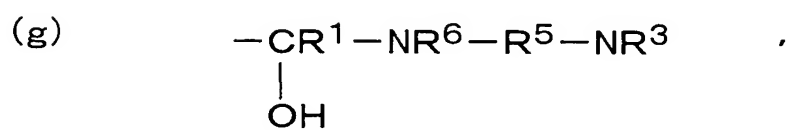
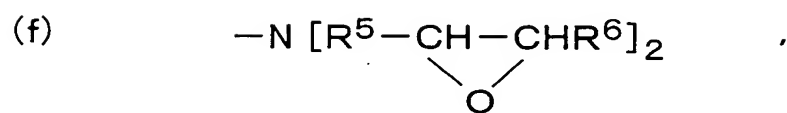
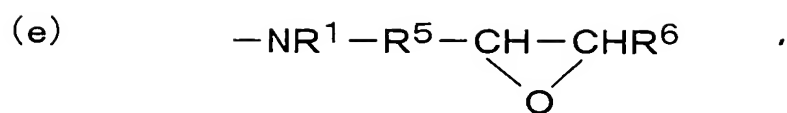
functional group-containing modifier group, wherein
 said second-order modifier has a functional group which
 is reactive to said functional group of said first
 -order modifier group (γ) of said first-order modified
 5 polymer, and wherein said second-order modifier is used
 in an amount of 0.3 to 10 moles, relative to one
 equivalent of said functional group of said first-order
 modifier group (γ) of said first-order modified polymer,
 thereby obtaining a second-order modified polymer,

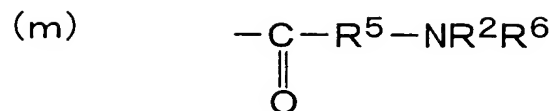
10 wherein said functional group-containing
 first-order modifier group (γ) of said first-
 order modified polymer comprises at least one
 functional group represented by a formula se-
 lected from the group consisting of the follow-
 15 ing formulae (a) to (m):



20







wherein, in the formulae (a) to (m):

5 N represents a nitrogen atom, Si represents a silicon atom, O represents an oxygen atom, C represents a carbon atom, and H represents a hydrogen atom,

10 each of R^1 to R^4 independently represents a hydrogen atom or a C_1 - C_{24} hydrocarbon group which optionally has at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol group and a C_1 - C_{24} alkoxysilane group,

15 each R^5 independently represents a C_1 - C_{48} hydrocarbon group and optionally, independently has at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol group and a C_1 - C_{24} alkoxysilane group,

20 each R^6 independently represents a hydrogen atom or a C_1 - C_8 alkyl group,

25 wherein each of R^1 to R^5 optionally, in-

dependently has bonded thereto at least one atom selected from the group consisting of an oxygen atom, a nitrogen atom, a sulfur atom and a silicon atom, said at least one atom being present in a linkage other than a hydroxyl group, an epoxy group, an amino group, a silanol group and an alkoxysilane group.

10 25. A filler-containing modified polymer composition comprising:

100 parts by weight of (A-2) the second-order modified polymer of any one of claims 20 to 23,

15 0.5 to 300 parts by weight of (B) a reinforcing filler.

26. The filler-containing modified polymer composition according to claim 25, wherein said reinforcing filler (B) is at least one member selected from the group consisting of a silica inorganic filler, a metal oxide, a metal hydroxide and carbon.

27. A crosslinked, filler-containing modified polymer composition obtained by subjecting the filler -containing modified polymer composition of claim 25 or

26 to a crosslinking reaction in the presence of a vulcanizing agent.

28. A modified polymer composition comprising:

5 1 to 99 parts by weight, relative to 100 parts by weight of the total of components (A-2) and (D), of (A-2) the second-order modified polymer of any one of claims 20 to 23, and

 99 to 1 part by weight, relative to 100 parts by weight of the total of components (A-2) and (D), of (D) at least one polymer selected from the group consisting of a thermoplastic resin other than said second-order modified polymer (A-2) and a rubbery polymer other than said second-order modified polymer (A-2).

15

29. The modified polymer composition according to claim 28, wherein said thermoplastic resin in component (D) comprises at least one member selected from the group consisting of a polyester resin, a polyamide resin, a polycarbonate resin, a polyurethane resin, a polyphenylene ether resin and a polyoxymethylene resin each of which contains at least one functional group selected from the group consisting of an acid anhydride group, a carboxyl group, a hydroxyl group, an epoxy group, an amino group, a silanol group and an alkoxy-

20

25

silane group.

30. A crosslinked, modified polymer composition obtained by subjecting the modified polymer composition
5 of any one of claim 28 or 29 to melt-kneading in the presence of a vulcanizing agent.

31. An adhesive composition comprising:
100 parts by weight of (A-2) the second-order
10 modified polymer of any one of claims 20 to 23, and
20 to 400 parts by weight of (E) a tackifier.

32. An asphalt composition comprising:
0.5 to 50 parts by weight of (A-2) the second
15 -order modified polymer of any one of claims 20 to 23,
and
100 parts by weight of (F) an asphalt.

33. A styrene resin composition obtained by subjecting
20 a raw material mixture to radical polymerization, said
raw material mixture comprising:

2 to 30 parts by weight, relative to 100 parts by
weight of the total of components (A-2) and (G), of
(A-2) the second-order modified polymer of any one of
25 claims 20 to 23, and

98 to 70 parts by weight, relative to 100 parts by weight of the total of components (A-2) and (G), of (G) a vinyl aromatic hydrocarbon monomer or a mixture of a vinyl aromatic hydrocarbon monomer and a comonomer copolymerizable with said vinyl aromatic hydrocarbon monomer.

34. The styrene resin composition according to claim 33, wherein said raw material mixture further comprises 0.5 to 300 parts by weight, relative to 100 parts by weight of component (A-2), of (B) a reinforcing filler.

35. The styrene resin composition according to claim 34, wherein said reinforcing filler (B) is at least one member selected from the group consisting of a silica inorganic filler, a metal oxide, a metal hydroxide and carbon.

36. A filler-containing modified polymer composition comprising:

100 parts by weight of (A-3) a first-order modified polymer comprising:

5 (β) a base polymer which is unhydrogenated or at least partially hydrogenated and which is at least one member selected from the group consisting of the following polymers (β -1) to (β -3):

10 (β -1) a conjugated diene polymer comprising conjugated diene monomer units,

15 (β -2) a copolymer comprising conjugated diene monomer units and vinyl aromatic hydrocarbon monomer units, said copolymer having no or at least one polymer block (H) of said vinyl aromatic hydrocarbon monomer units, wherein said copolymer has a vinyl aromatic hydrocarbon block ratio of from 0 to less than 50 % by weight, said vinyl aromatic hydrocarbon block ratio being defined as the percent by weight of
20 the vinyl aromatic hydrocarbon monomer units contained in said at least one polymer block (H) of said vinyl aromatic hydrocarbon monomer units, based on the total weight of vinyl aromatic hydrocarbon monomer units contained in
25 said copolymer as in the unhydrogenated state,

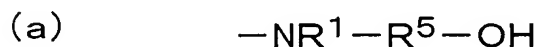
and

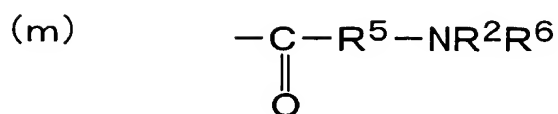
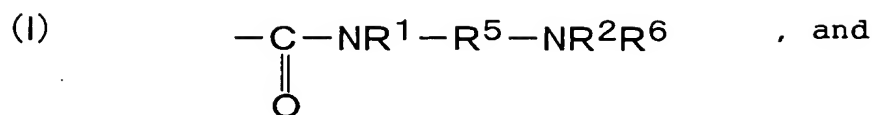
(β -3) a vinyl aromatic hydrocarbon polymer comprising vinyl aromatic hydrocarbon monomer units, and

5 (γ) a functional group-containing first-order modifier group bonded to said base polymer (β),
0.5 to 300 parts by weight of (B) a reinforcing filler, and

0.01 to 20 parts by weight of (C) a second-order
10 modifier having a functional group which is reactive to said functional group of said first-order modifier group (γ) of said first-order modified polymer (A-3), wherein said second-order modifier (C) is at least one member selected from the group consisting of a func-
15 tional monomer and a functional oligomer,

wherein said functional group-containing first-order modifier group (γ) of said first-order modified polymer (A-3) comprises at least one functional group represented by a
20 formula selected from the group consisting of the following formulae (a) to (m):





wherein, in the formulae (a) to (m):

N represents a nitrogen atom, Si represents a silicon atom, O represents an oxygen atom, C represents a carbon atom, and H represents a hydrogen atom,

each of R^1 to R^4 independently represents a hydrogen atom or a $\text{C}_1\text{-C}_{24}$ hydrocarbon group which optionally has at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol group and a $\text{C}_1\text{-C}_{24}$ alkoxysilane group,

each R^5 independently represents a $\text{C}_1\text{-C}_{48}$ hydrocarbon group and optionally, independently has at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol group and a $\text{C}_1\text{-C}_{24}$ alkoxysilane group,

each R⁶ independently represents a hydrogen atom or a C₁-C₈ alkyl group,

wherein each of R¹ to R⁵ optionally, independently has bonded thereto at least one atom selected from the group consisting of an oxygen atom, a nitrogen atom, a sulfur atom and a silicon atom, said at least one atom being present in a linkage other than a hydroxyl group, an epoxy group, an amino group, a silanol group and an alkoxysilane group.

37. The filler-containing modified polymer composition according to claim 36, wherein said reinforcing filler (B) is at least one member selected from the group consisting of a silica inorganic filler, a metal oxide, a metal hydroxide and carbon.

38. A crosslinked, filler-containing modified polymer composition obtained by subjecting the filler-containing modified polymer composition of claim 36 or 37 to a crosslinking reaction in the presence of a vulcanizing agent.

39. A modified polymer composition comprising:

1 to 99 parts by weight, relative to 100 parts by weight of the total of components (A-3) and (D), of (A-3) a first-order modified polymer comprising:

(β) a base polymer which is unhydrogenated or at least partially hydrogenated and which is at least one member selected from the group consisting of the following polymers (β -1) to (β -3):

(β -1) a conjugated diene polymer comprising conjugated diene monomer units,

(β -2) a copolymer comprising conjugated diene monomer units and vinyl aromatic hydrocarbon monomer units, said copolymer having no or at least one polymer block (H) of said vinyl aromatic hydrocarbon monomer units, wherein said copolymer has a vinyl aromatic hydrocarbon block ratio of from 0 to less than 50 % by weight, said vinyl aromatic hydrocarbon block ratio being defined as the percent by weight of the vinyl aromatic hydrocarbon monomer units contained in said at least one polymer block (H) of said vinyl aromatic hydrocarbon monomer units, based on the total weight of vinyl aromatic hydrocarbon monomer units contained in said copolymer as in the unhydrogenated state, and

(β -3) a vinyl aromatic hydrocarbon polymer comprising vinyl aromatic hydrocarbon monomer units, and

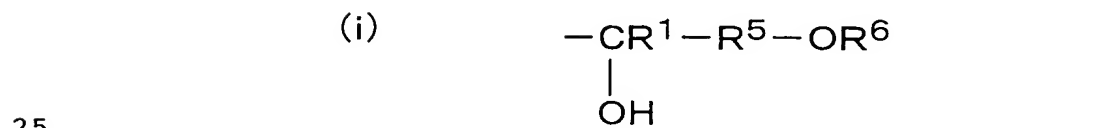
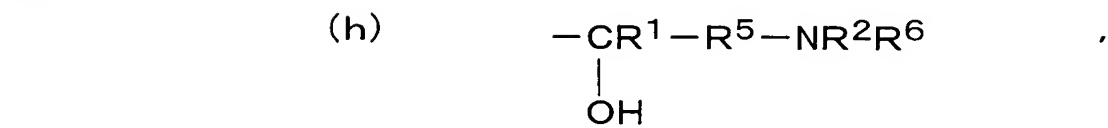
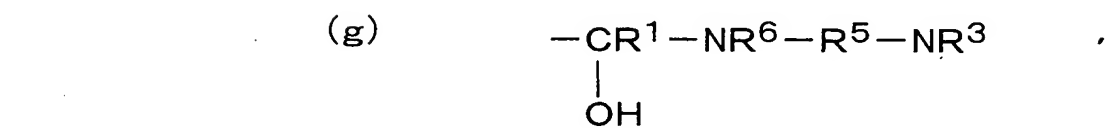
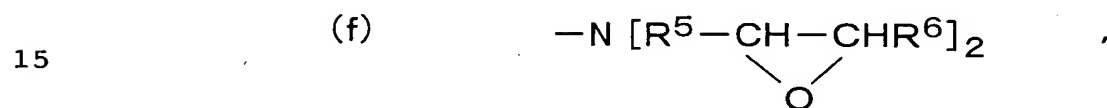
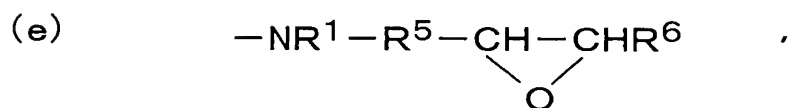
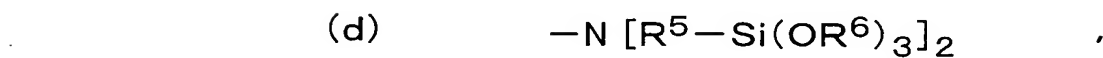
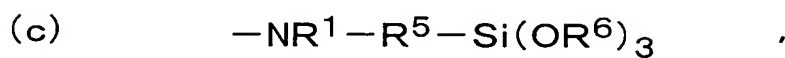
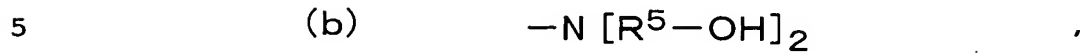
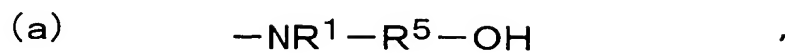
5 (γ) a functional group-containing first-order modifier group bonded to said base polymer (β),

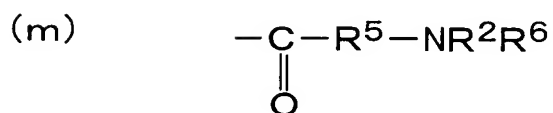
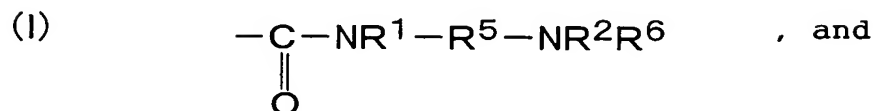
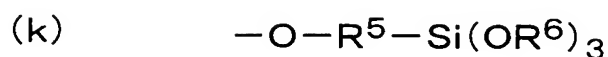
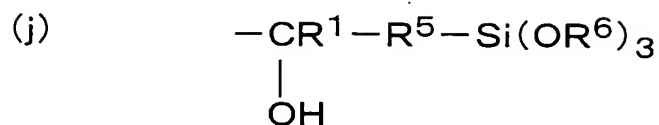
99 to 1 part by weight, relative to 100 parts by weight of the total of components (A-3) and (D), of (D) at least one polymer selected from the group consisting of a thermoplastic resin other than said first-order modified polymer (A-3) and a rubbery polymer other than
10 said first-order modified polymer (A-3), and

0.01 to 20 parts by weight, relative to 100 parts by weight of the total of components (A-3) and (D), of (C) a second-order modifier having a functional group
15 which is reactive to said functional group of said first-order modifier group (γ) of said first-order modified polymer (A-3), wherein said second-order modifier (C) is at least one member selected from the group consisting of a functional monomer and a functional
20 oligomer,

wherein said functional group-containing first-order modifier group (γ) of said first-order modified polymer (A-3) comprises at least one functional group represented by a
25 formula selected from the group consisting of

the following formulae (a) to (m):





wherein, in the formulae (a) to (m):

N represents a nitrogen atom, Si represents a silicon atom, O represents an oxygen atom, C represents a carbon atom, and H represents a hydrogen atom,

each of R^1 to R^4 independently represents a hydrogen atom or a C_1 - C_{24} hydrocarbon group which optionally has at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol group and a C_1 - C_{24} alkoxysilane group,

each R^5 independently represents a C_1 - C_{48} hydrocarbon group and optionally, inde-

pendently has at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol group and a C₁-C₂₄ alkoxy silane group,

each R⁶ independently represents a hydrogen atom or a C₁-C₈ alkyl group,

wherein each of R¹ to R⁵ optionally, independently has bonded thereto at least one atom selected from the group consisting of an oxygen atom, a nitrogen atom, a sulfur atom and a silicon atom, said at least one atom being present in a linkage other than a hydroxyl group, an epoxy group, an amino group, a silanol group and an alkoxy silane group.

40. The modified polymer composition according to claim 39, wherein said thermoplastic resin in component (D) comprises at least one member selected from the group consisting of a polyester resin, a polyamide resin, a polycarbonate resin, a polyurethane resin, a polyphenylene ether resin and a polyoxymethylene resin each of which contains at least one functional group selected from the group consisting of an acid anhydride

group, a carboxyl group, a hydroxyl group, an epoxy group, an amino group, a silanol group and an alkoxy-silane group.

5 41. A crosslinked, modified polymer composition obtained by subjecting the modified polymer composition of claim 39 or 40 to melt-kneading in the presence of a vulcanizing agent.

10 42. An adhesive composition comprising:
100 parts by weight of (A-3) a first-order modified polymer comprising:

(β) a base polymer which is unhydrogenated or at least partially hydrogenated and which is at
15 least one member selected from the group consisting of the following polymers (β -1) to (β -3):

(β -1) a conjugated diene polymer comprising conjugated diene monomer units,

(β -2) a copolymer comprising conjugated di-
20 ene monomer units and vinyl aromatic hydrocarbon monomer units, said copolymer having no or at least one polymer block (H) of said vinyl aromatic hydrocarbon monomer units, wherein said copolymer has a vinyl aromatic hydrocarbon
25 block ratio of from 0 to less than 50 % by

weight, said vinyl aromatic hydrocarbon block ratio being defined as the percent by weight of the vinyl aromatic hydrocarbon monomer units contained in said at least one polymer block (H) of said vinyl aromatic hydrocarbon monomer units, based on the total weight of vinyl aromatic hydrocarbon monomer units contained in said copolymer as in the unhydrogenated state, and

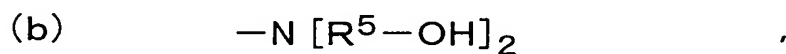
(β -3) a vinyl aromatic hydrocarbon polymer comprising vinyl aromatic hydrocarbon monomer units, and

(γ) a functional group-containing first-order modifier group bonded to said base polymer (β),
20 to 400 parts by weight of (E) a tackifier, and
0.01 to 20 parts by weight of (C) a second-order modifier having a functional group which is reactive to said functional group of said first-order modifier group (γ) of said first-order modified polymer (A-3),
wherein said second-order modifier (C) is at least one member selected from the group consisting of a functional monomer and a functional oligomer,

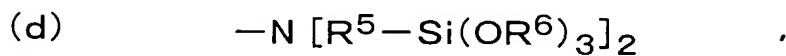
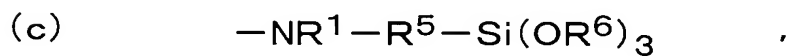
wherein said functional group-containing first-order modifier group (γ) of said first-order modified polymer (A-3) comprises at least

one functional group represented by a formula selected from the group consisting of the following formulae (a) to (m):

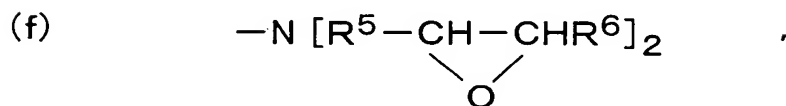
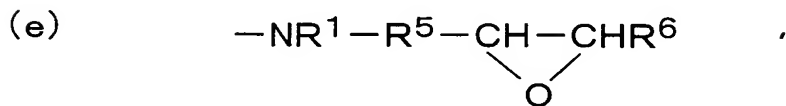
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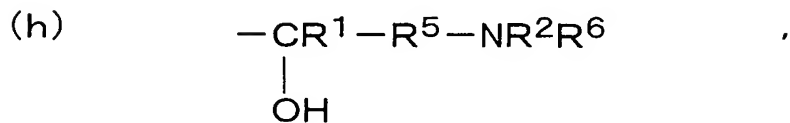
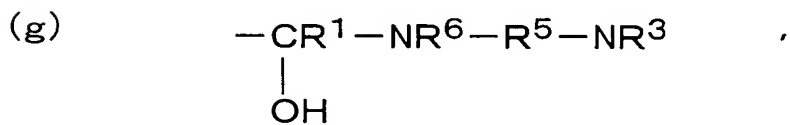
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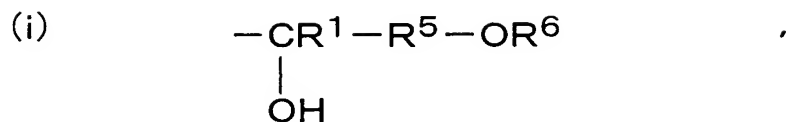


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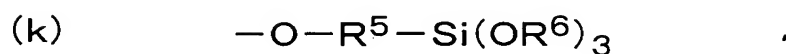
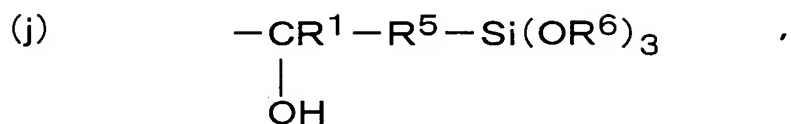


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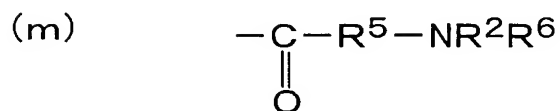
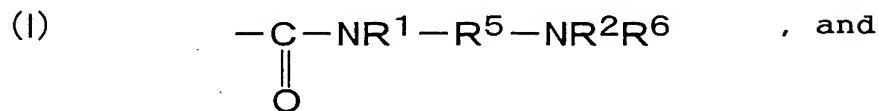




5



10



15

wherein, in the formulae (a) to (m):

N represents a nitrogen atom, Si represents a silicon atom, O represents an oxygen atom, C represents a carbon atom, and H represents a hydrogen atom,

20

each of R^1 to R^4 independently represents a hydrogen atom or a C_1 - C_{24} hydrocarbon group which optionally has at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol

25

group and a C₁-C₂₄ alkoxysilane group,

each R⁵ independently represents a C₁-C₄₈ hydrocarbon group and optionally, independently has at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol group and a C₁-C₂₄ alkoxysilane group,

each R⁶ independently represents a hydrogen atom or a C₁-C₈ alkyl group,

wherein each of R¹ to R⁵ optionally, independently has bonded thereto at least one atom selected from the group consisting of an oxygen atom, a nitrogen atom, a sulfur atom and a silicon atom, said at least one atom being present in a linkage other than a hydroxyl group, an epoxy group, an amino group, a silanol group and an alkoxysilane group.

43. An asphalt composition comprising:

0.5 to 50 parts by weight of (A-3) a first-order modified polymer comprising:

(β) a base polymer which is unhydrogenated or at least partially hydrogenated and which is at

least one member selected from the group consisting of the following polymers (β -1) to (β -3):

(β -1) a conjugated diene polymer comprising conjugated diene monomer units,

5 (β -2) a copolymer comprising conjugated diene monomer units and vinyl aromatic hydrocarbon monomer units, said copolymer having no or at least one polymer block (H) of said vinyl aromatic hydrocarbon monomer units, wherein
10 said copolymer has a vinyl aromatic hydrocarbon block ratio of from 0 to less than 50 % by weight, said vinyl aromatic hydrocarbon block ratio being defined as the percent by weight of the vinyl aromatic hydrocarbon monomer units
15 contained in said at least one polymer block (H) of said vinyl aromatic hydrocarbon monomer units, based on the total weight of vinyl aromatic hydrocarbon monomer units contained in said copolymer as in the unhydrogenated state,
20 and

(β -3) a vinyl aromatic hydrocarbon polymer comprising vinyl aromatic hydrocarbon monomer units, and

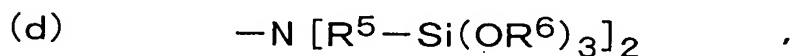
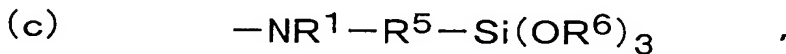
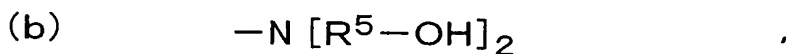
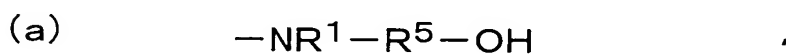
(γ) a functional group-containing first-order
25 modifier group bonded to said base polymer (β),

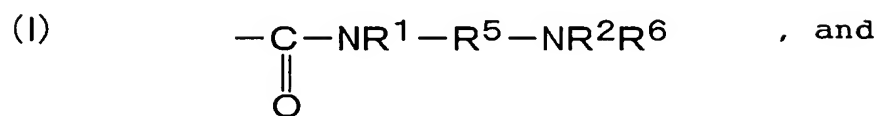
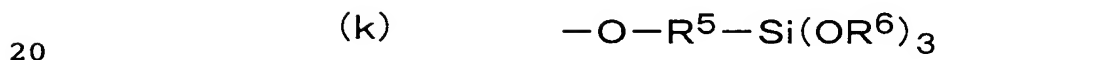
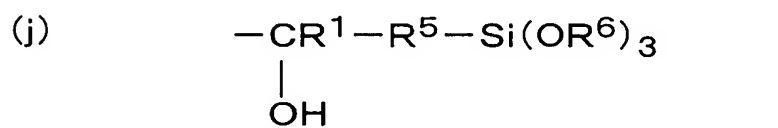
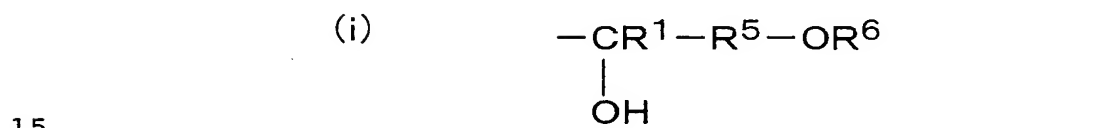
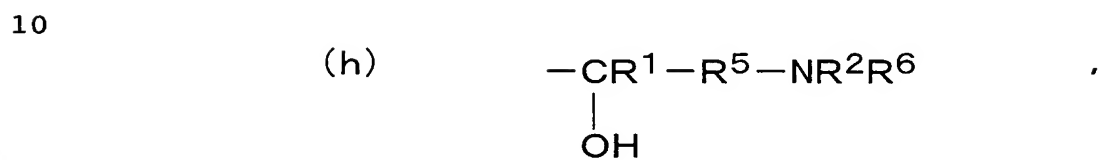
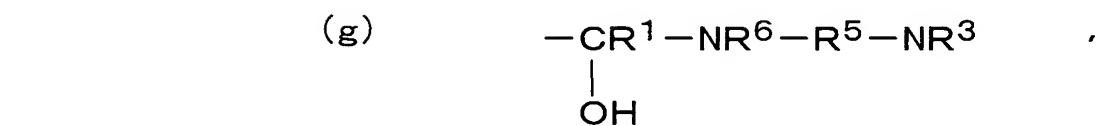
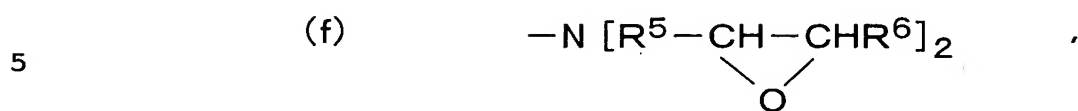
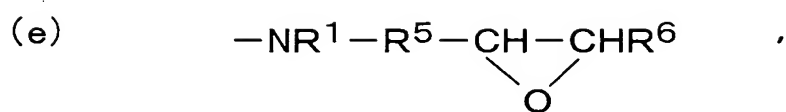
100 parts by weight of (F) an asphalt, and

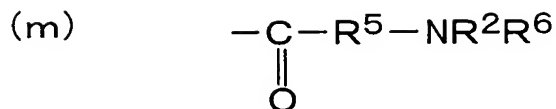
0.01 to 20 parts by weight of (C) a second-order modifier having a functional group which is reactive to said functional group of said first-order modifier

group (γ) of said first-order modified polymer (A-3), wherein said second-order modifier (C) is at least one member selected from the group consisting of a functional monomer and a functional oligomer,

wherein said functional group-containing first-order modifier group (γ) of said first-order modified polymer (A-3) comprises at least one functional group represented by a formula selected from the group consisting of the following formulae (a) to (m):







wherein, in the formulae (a) to (m):

5 N represents a nitrogen atom, Si represents a silicon atom, O represents an oxygen atom, C represents a carbon atom, and H represents a hydrogen atom,

10 each of R^1 to R^4 independently represents a hydrogen atom or a C_1 - C_{24} hydrocarbon group which optionally has at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol group and a C_1 - C_{24} alkoxysilane group,

15 each R^5 independently represents a C_1 - C_{48} hydrocarbon group and optionally, independently has at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol group and a C_1 - C_{24} alkoxysilane group,

20 each R^6 independently represents a hydrogen atom or a C_1 - C_8 alkyl group,

25 wherein each of R^1 to R^5 optionally, in-

dependently has bonded thereto at least one atom selected from the group consisting of an oxygen atom, a nitrogen atom, a sulfur atom and a silicon atom, said at least one atom being present in a linkage other than a hydroxyl group, an epoxy group, an amino group, a silanol group and an alkoxysilane group.

44. A styrene resin composition obtained by subjecting a raw material mixture to radical polymerization, said raw material mixture comprising:

2 to 30 parts by weight, relative to 100 parts by weight of the total of components (A-3) and (G), of

(A-3) a first-order modified polymer comprising:

(β) a base polymer which is unhydrogenated or at least partially hydrogenated and which is at least one member selected from the group consisting of the following polymers (β -1) to (β -3):

(β -1) a conjugated diene polymer comprising conjugated diene monomer units,

(β -2) a copolymer comprising conjugated diene monomer units and vinyl aromatic hydrocarbon monomer units, said copolymer having no or at least one polymer block (H) of said vinyl

aromatic hydrocarbon monomer units, wherein
said copolymer has a vinyl aromatic hydrocarbon
block ratio of from 0 to less than 50 % by
weight, said vinyl aromatic hydrocarbon block
5 ratio being defined as the percent by weight of
the vinyl aromatic hydrocarbon monomer units
contained in said at least one polymer block
(H) of said vinyl aromatic hydrocarbon monomer
units, based on the total weight of vinyl aro-
10 matic hydrocarbon monomer units contained in
said copolymer as in the unhydrogenated state,
and

(β -3) a vinyl aromatic hydrocarbon polymer
comprising vinyl aromatic hydrocarbon monomer
15 units, and

(γ) a functional group-containing first-order
modifier group bonded to said base polymer (β),

98 to 70 parts by weight, relative to 100 parts by
weight of the total of components (A-3) and (G), of (G)
20 a vinyl aromatic hydrocarbon monomer or a mixture of a
vinyl aromatic hydrocarbon monomer and a comonomer co-
polymerizable with said vinyl aromatic hydrocarbon
monomer, and

0.01 to 20 parts by weight, relative to 100 parts
25 by weight of the total of components (A-3) and (G), of

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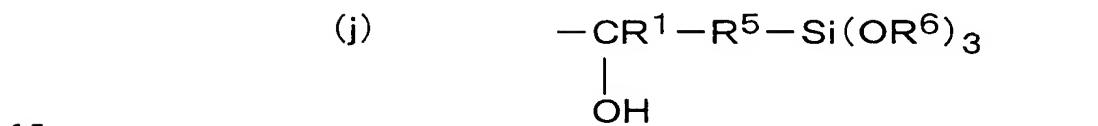
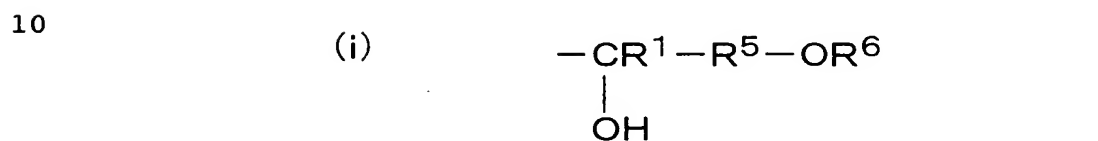
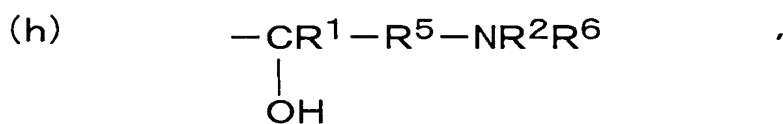
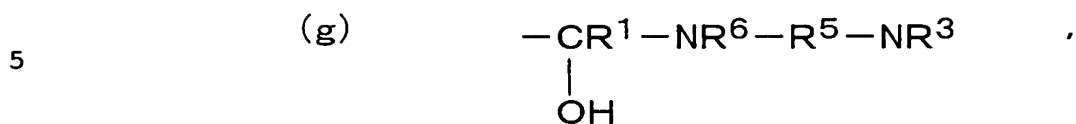
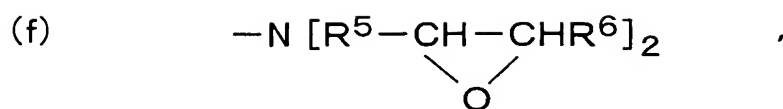
(a) $-NR^1-R^5-OH$

(b) $-N [R^5-OH]_2$

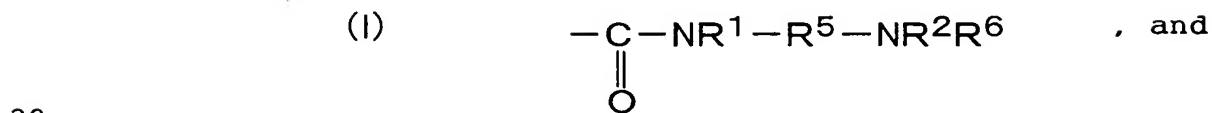
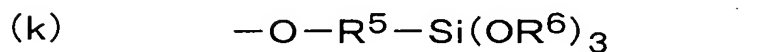
(c) $-NR^1-R^5-Si(OR^6)_3$

(d) $-N [R^5-Si(OR^6)_3]_2$

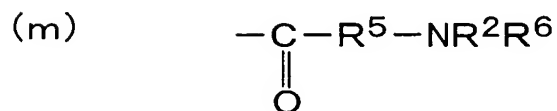
(e)
$$-NR^1-R^5-\underset{\text{O}}{\underset{|}{\text{CH}}}-\text{CHR}^6$$



15



20



wherein, in the formulae (a) to (m):

25

N represents a nitrogen atom, Si repre-

sents a silicon atom, O represents an oxygen atom, C represents a carbon atom, and H represents a hydrogen atom,

5 each of R^1 to R^4 independently represents a hydrogen atom or a C_1 - C_{24} hydrocarbon group which optionally has at least one functional group selected from the group consisting of a hydroxyl group, an epoxy group, an amino group, a silanol
10 group and a C_1 - C_{24} alkoxysilane group,

each R^5 independently represents a C_1 - C_{48} hydrocarbon group and optionally, independently has at least one functional group selected from the group consisting
15 of a hydroxyl group, an epoxy group, an amino group, a silanol group and a C_1 - C_{24} alkoxysilane group,

each R^6 independently represents a hydrogen atom or a C_1 - C_8 alkyl group,

20 wherein each of R^1 to R^5 optionally, independently has bonded thereto at least one atom selected from the group consisting of an oxygen atom, a nitrogen atom, a sulfur atom and a silicon atom, said at
25 least one atom being present in a linkage

other than a hydroxyl group, an epoxy group, an amino group, a silanol group and an alkoxysilane group.

- 5 45. The styrene resin composition according to claim 44, wherein said raw material mixture further comprises 0.5 to 300 parts by weight, relative to 100 parts by weight of component (A-3), of (B) a reinforcing filler.
- 10 46. The styrene resin composition according to claim 45, wherein said reinforcing filler (B) is at least one member selected from the group consisting of a silica inorganic filler, a metal oxide, a metal hydroxide and carbon.